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RECRYSTALLIZATION KINETICS OF NANOSTRUCUTRED COPPER PROCESSED BY DYNAMIC PLASTIC DEFORMATION

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ABSTRACT

The recrystallization kinetics of nanostructured copper samples processed by dynamic plastic deformation was investigated by electron backscatter diffraction. It was found that the evolution of the recrystallized volume fraction as a function of annealing time has a very low slope (n=0.37) when plotted as an Avrami plot. Various reasons for such a low slope are discussed, including possible recrystallization during storage of samples, and the heterogeneous recrystallization kinetics. The effects of heterogeneous recrystallization kinetics are illustrated by a simplified model with a fast and a slowly recrystallizing region.

1. INTRODUCTION

Dynamic plastic deformation (DPD) is an effective method in producing nanostructured materials (Zhao, Tao, Guo, Lu and Lu 2005), which can greatly improve the strength of the material. Thermal stability is however always an important issue for nanostructured materials produced by high strain deformation. In some cases, recrystallization can happen at a low temperature, and thus even room temperature storage may modify the microstructure (Mishin and Godfrey 2008). Recrystallization and partial recrystallization typically reduce the strength of nanometals, but a partly recrystallized microstructure may improve the ductility of the material (Wang, Chen, Zhou and Ma 2002). Nevertheless, the improvement in ductility by introducing recrystallized grains varies for samples produced by different deformation techniques. The uniform tensile elongation has been observed to increase from a few percent to around 30% by introducing 25% volume fraction of recrystallized grains in commercial purity copper processed by rolling at liquid nitrogen temperature to 95% thickness reduction (Wang, et

al. 2002), while pure copper deformed by DPD at liquid nitrogen temperature did not obtain any obvious ductility improvement before the recrystallized volume fraction was as high as 80% (Li, Zhang, Tao and Lu 2008). It is thus considered important to understand in more detail the recrystallization process and the microstructural evolution of the nanostructured samples during heat treatment. In this paper, the overall recrystallization kinetics and its relationship with the heterogeneous deformation structure in samples processed by DPD are investigated.

2. EXPERIMENTAL DETAILS

Copper of 99.995% purity and an initial grain size of 200 μm was used. A cylinder 9 mm in diameter and 26 mm in height was processed by DPD at liquid nitrogen temperature at IMR Shenyang. The cylinder was deformed by 17 impacts. The height reduction at each step varied from 2 mm in the beginning to 0.1 mm at the end. The final height of the sample was 3.3 mm, corresponding to an equivalent strain of 2.0. Annealing was performed at 120 °C for various time periods from 2 minutes to 4 h. Cross sections containing the compression axis were characterized by electron backscatter diffraction (EBSD) with a step size of 0.5 μm. A typical scan area covered 250 μm along the compression axis, and 150 μm along the other direction. For each annealing time period, 2 to 7 scans were performed; the microstructural parameters were averaged over all scans for each sample.

For identification of recrystallized grains from the EBSD data, the so called DRG program was used (Wu and Juul Jensen 2008). The definition of recrystallized grains is as follows 1) the pixel to pixel misorientation inside a recrystallized grain should be less than 1°; 2) the minimum size defined as the equivalent circular diameter (ECD) is 1.5 μm; 3) at least part of the boundaries surrounding the grain should be high angle boundaries with a misorientation larger than 15°. Twin boundaries developed inside recrystallized grains are excluded in the grain recognition, which means that a grain and its twin parts are considered as one grain. The volume fraction of recrystallized material (V_V) within each sample is calculated as an area fraction.

3. RESULTS

The recrystallized volume fraction V_V is plotted as a function of annealing time in Fig. 1a. The V_V is approximately 30% after only 2 minutes annealing at 120°C. The recrystallized volume fraction increases slowly to approximately 60% after 30 minutes annealing, and 80% after 4h annealing. The error bars show the standard deviations of V_V from different scan areas within each sample. The standard deviations of V_V are large, especially in the beginning of annealing. The standard deviation of V_V for the 2 minutes annealed sample is 28%. Among the 7 maps characterized for the 2 minutes annealed sample, the minimum V_V is 1.6%, while the maximum is 69%. The standard deviation of V_V decreases slightly after longer time annealing, nevertheless it is still as high as 11% after 4 h annealing. This large scatter in V_V is a consequence of the heterogeneous recrystallization occurring in these samples (Lin, Zhang, Tao, Pantleon and Juul Jensen 2012).

The evolution of V_V with time t is often expressed by the JMAK equation:

$$V_V = 1 - \exp(-kt^n) \quad (1)$$

which can also be written as $\log(-\ln(1 - V_V)) = n \cdot \log(t) + \log(k)$ (Avrami 1939). The data of Fig. 1a can thus be plotted as $-\ln(1 - V_V)$ as a function of time on a log-log scale, which is usually termed an Avrami plot (see Fig. 1b). The data points fit nicely a straight line. The slope

of the line, which corresponds to the exponent n in the eq. (1) is 0.37.

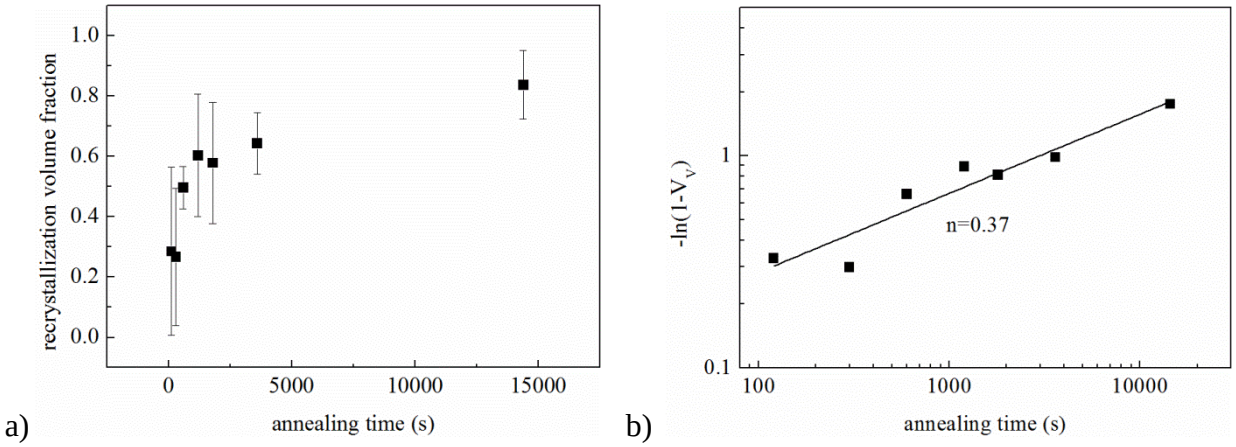


Fig. 1. a) Evolution of the recrystallized volume fraction as a function of annealing time and b) Avrami plot of the same data as in a).

The microstructure after 1h annealing is shown at Fig. 2a and b. Fig. 2a is an orientation map reconstructured from EBSD data colored according to the crystallographic direction of the compression axis. Fig. 2b highlights the recrystallized grains using random colors for individual recrystallized grains and the deformed matrix is shown in black. The area fraction of recrystallized grains in this map is approximately 50%. The majority of recrystallized grains cluster in 3 bands, perpendicular to the compression axis. The band to the left (band 1) is composed of a large number of small grains while the band to the right (band 3) is a mixture of small grains and a few very large grains. The band in the middle (band 2) also contains small and big grains, while the big grains in band 2 are not as big as in band 3. A high density of twin boundaries appears in most of the recrystallized grains. It is however interesting to notice that the area of twin parts in the few large grains are very limited. The remaining deformed matrix is divided into 3 bands by the bands of recrystallized grains, in which only a few recrystallized grains lie along directions about 60° to the compression axis.

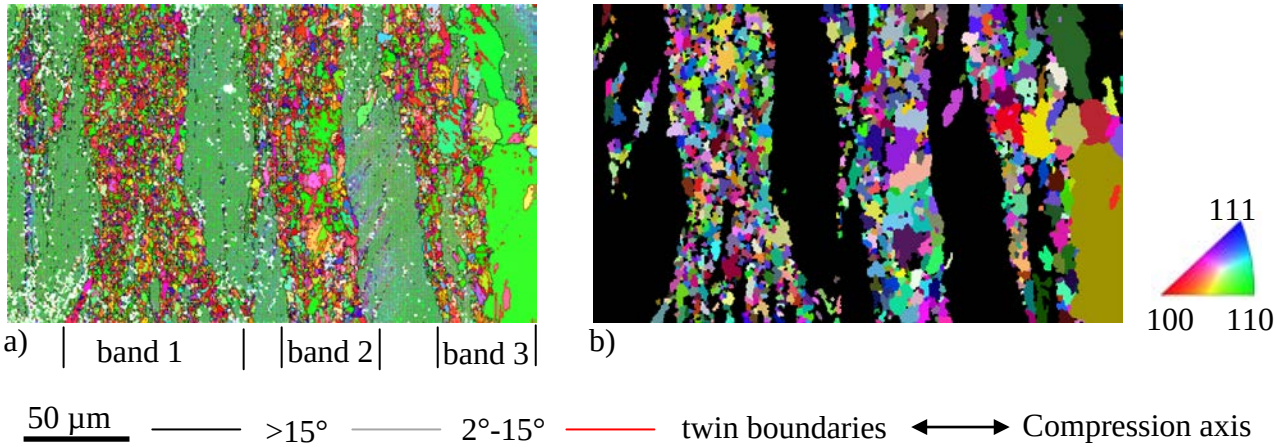
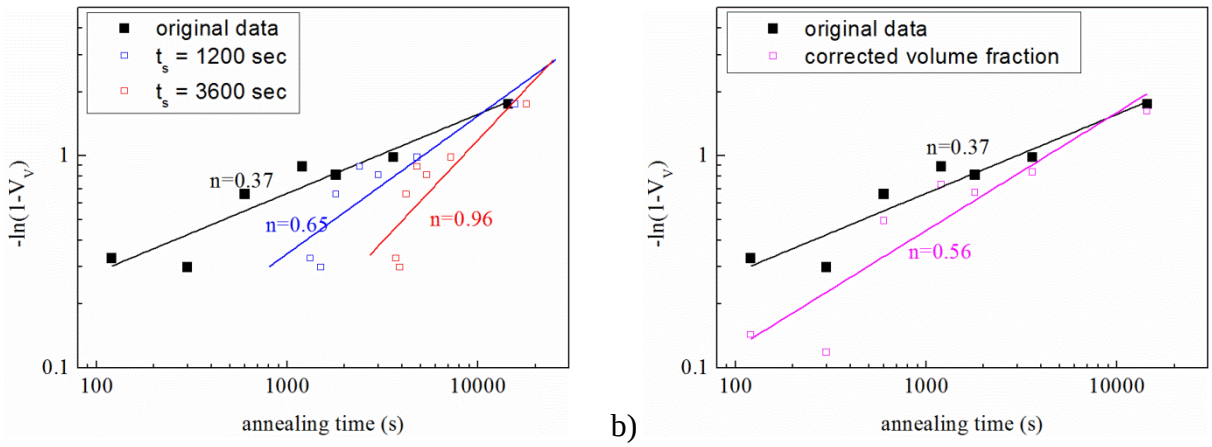


Fig. 2. a) Orientation map showing the microstructure after 1 h annealing at 120 °C; colors represent the crystallographic direction of the compression axis; points not indexed by EBSD are in white b) same map as a) with recrystallized grains highlighted in random colors.

4. DISCUSSION

The classical recrystallization kinetics model is the JMAK model given in eq. (1). This model is based on the assumption that nucleation sites are randomly distributed and all grains grow with one constant growth rate. The JMAK model predicts an exponent n of 3 and 4 for 3D growth with site-saturated nucleation and a constant nucleation rate, respectively. This model is however too simplified to be compared with many recrystallization processes. For example, a non-random distribution of nucleation sites is often observed, which changes the recrystallization kinetics due to early impingement of recrystallized grains (Vandermeer 2005; Storm and Juul Jensen 2009). Moreover, the growth rates of grains are often observed to vary over time and from grain to grain (Godiksen, Schmidt and Juul Jensen 2007; Lauridsen, Poulsen, Nielsen and Juul Jensen 2003) and even different parts of a grain boundary may migrate differently (Zhang, Godfrey, Liu, Liu and Juul Jensen 2009), which also affects the recrystallization kinetics. The exponent n is thus often found to be less than 3. There have been numerous cases in which exponents n in the order of 1 have been found (Humphreys and Hatherly 2004). The exponent n of 0.37 in the present case is however even lower.

One may speculate that a possible reason for such a low exponent could be recrystallization during storage of the materials. The present DPD samples were kept at -18°C for around 6 months plus some days at room temperature during transportation from China to Denmark and sample preparation. It is possible that part of the 30% volume fraction of recrystallized grains after 2 minutes annealing existed before furnace annealing, and formed during the storage and/or sample preparation. The recrystallization time t should then be $t+t_s$, where t_s is the storage time converted into an equivalent time at 120°C. If t_s is set to 20 minutes and 1 h, the fitted exponent n increases from 0.37 to 0.65 and 0.96 respectively (Fig. 3a). However even 20 minutes may be an overestimation, as the reported recrystallization activation energy varies from 58 kJ to 170 kJ (Jagle and Mittemeijer 2011), and even if the lowest activation energy is taken, 6 months storage at -18°C corresponds to only 18 minutes annealing at 120 °C. The DPD samples were recently checked after 18 months storage, and the recrystallization volume faction was found to be 17.5% ± 16.7%. This may be incorporated in the analysis as $V_V' = (V_V - V_{V0})/(1 - V_{V0})$, where V_{V0} is the recrystallized volume fraction after storage. The Avrami plot of V_V' is shown in Fig. 3b, and the exponent n equals 0.56, i.e. the exponent n is still very low.



a) b)

Fig. 3. Avrami plot of the recrystallized volume fraction a) by assuming the low temperature storage time corresponds to 20 minutes and 1 h annealing time t_s at 120 °C; b) corrected as $V_V' = (V_V - V_{V0})/(1 - V_{V0})$, where V_{V0} is the recrystallized volume fraction after storage.

Another explanation for the low exponent n could be the heterogeneous distribution of stored energies, and thus different recrystallization kinetics in different regions of the sample. The standard deviations of V_V shown as error bars in Fig. 2 are an obvious indication of different recrystallization kinetics at different places. After 2 minutes annealing, some regions (typically 150 µm by 250µm) have V_V>0.5, while some regions are almost not recrystallized at all. The microstructure in Fig. 2 clearly shows the heterogeneity of the recrystallization process. In the three bands of recrystallized grains, the recrystallization process has almost finished, while in other three bands, recrystallization has just begun. It is then reasonable to assume that at least two different types of regions exist in the sample, namely fast recrystallizing regions and slowly recrystallizing regions. The fast recrystallizing regions could relate to either a fast nucleation rate with a high density of nuclei (band 1) and/or fast growth rates of a few grains (band 3). In the slowly recrystallizing regions, the number of recrystallized grains seen after 1 h annealing is still very limited. It could be either that recrystallized grains have not yet formed in those regions, or that they are so small that only few of them appear in the characterized section.

To get an idea of how such heterogeneous recrystallization may affect the overall recrystallization kinetics, it is for simplicity assumed that the sample is composed of two regions A and B. Region A is a fast recrystallizing region, and region B is a slowly recrystallizing region. Each region has its own recrystallization kinetics as $V_{V_i} = 1 - \exp(-k_i t^{n_i})$. If the JMAK assumption is adopted, namely random, site saturated nucleation and 3D growth within each region, then the n_i is 3 for both regions. By further assuming that recrystallized grains only grow within their own region, the overall kinetics can then be expressed as:

$$V_V = xV_{V_A} + (1-x)V_{V_B} = x(1-\exp(-k_A t^3)) + (1-x)(1-\exp(-k_B t^3)) \qquad (2)$$

where x is the volume fraction of region A. The overall Avrami plot for eq. (2) is shown in Fig. 4 for various values of x (Fig. 4a) and k_A/k_B (Fig. 4b). The overall V_V line follows the kinetics line of the fast recrystallizing region in the beginning and the slowly recrystallizing region at the end, each with a slope close to 3. In between, the line bends over, and the slope at around V_V=0.5 may decrease to less than 1. Although this model is very simplified, it shows the possibility that the exponent n when obtained from relatively few experimental points at intermediate stages of recrystallization can be greatly reduced because of heterogeneity within the deformation matrix.

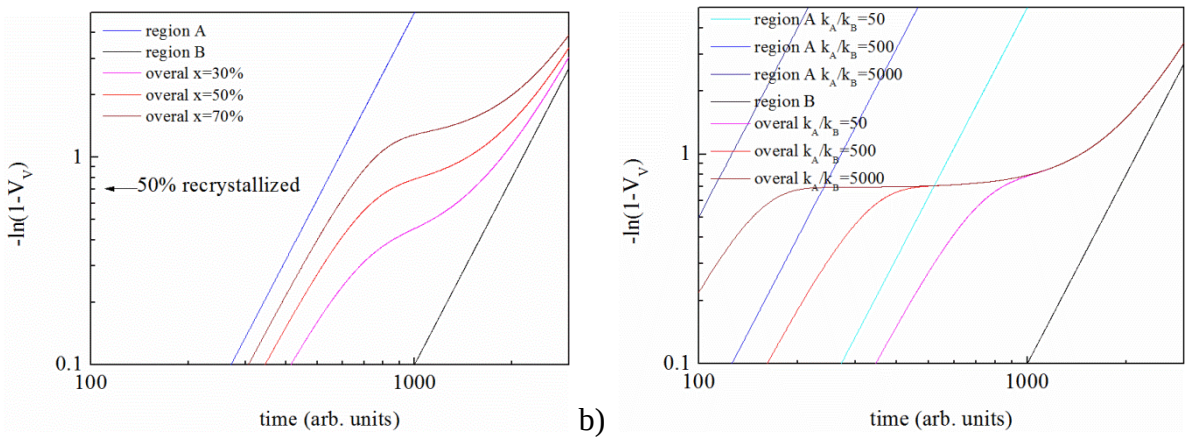


Fig. 4. Theoretical kinetics curves by assuming that there are two distinct regions in the sample, and each region follows JMAK recrystallization kinetics independently of each other with different k values. a) k_A/k_B=50, with different volume fraction of region A; b) 50% volume fraction of region A, with different values of k_A/k_B.

5. CONCLUSION

The recrystallization kinetics in pure copper deformed by dynamic plastic deformation (DPD) to a strain of 2.0 was characterized by EBSD and a very low kinetics exponent n=0.37 was observed. The partly recrystallized microstructure shows that the recrystallized grains cluster intensely in bands, which suggests that the recrystallization behavior is quite different in different regions of the sample. A simplified model assuming fast and slowly recrystallizing regions is proposed, leading to a very low overall exponent n at intermediate stages of recrystallization.

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